

Low temperature thermodynamic properties of Ce compounds tuned at $T_{ord} = 0$

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Based on specific heat (C_m) and entropy evaluation, different Ce magnetic phase diagrams can be recognized: I) with the entropy of the ordered phase (S_{MO}) decreasing with their order temperature (T_{ord}), which are the only candidates for quantum critical behavior since $S_{MO} \rightarrow 0$ as $T_{ord} \rightarrow 0$. II) with phase boundaries ending at a $T > 0$ critical point because their $C_m(T_{ord})$ jumps (ΔC_m) do not decrease sufficiently with T_{ord} producing a S_{MO} bottleneck, and III) those showing a transference of degrees of freedom to a non-magnetic component, with their ΔC_m vanishing at $T \gg 0$.

IV) There is also a group of Ce heavy fermions which do not order magnetically down to $T \cong 0$. These compounds are at the top of the $\lim_{T \rightarrow 0} \partial S_m / \partial T = C_m / T$ values because they collect very high density of low energy excitations. From the analysis of $C_m(T)/T$ and $S_m(T)$ results performed on selected Ce ternaries, a quantitative determination of an upper limit for the density of excitations is obtained, excluding any evidence of $C_m(T)/T$ divergency at $T \rightarrow 0$ in agreement with thermodynamic laws.

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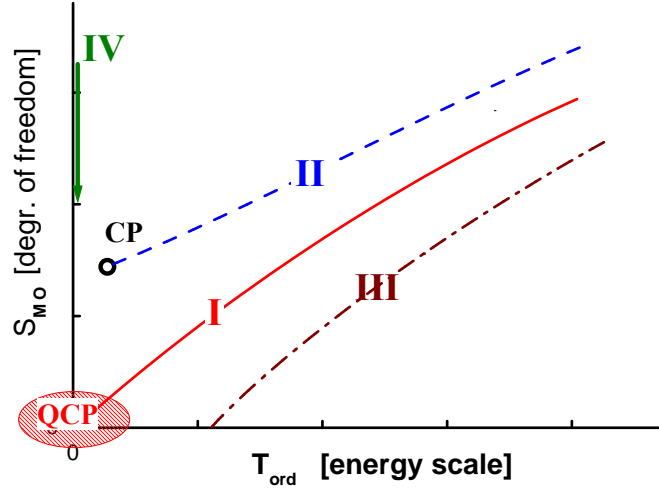


FIG. 1. Schematic comparison of three types of magnetic phase diagrams for the entropy of the ordered phase (S_{MO}) collected up to the transition at T_{ord} , after ref. [2]. The arrow label as 'IV' represents Ce compounds which do not show magnetic order down to $T \cong 0$ analyzed in this work.

I. INTRODUCTION.

Among the outstanding subjects of current investigations on strongly correlated electron systems are those related with quantum criticality (QC) [1]. Despite of the large amount of new intermetallic compounds claimed to be candidates to reach a quantum critical point (QCP) at $T = 0$, only a few of them were proved to reach that regime. Simple thermodynamic principles clearly establish the conditions for such scenario, which can be tested through the entropy (i.e the degrees of freedom) collected into the ordered phase S_{MO} up to the ordering temperature T_{ord} according to the $S_{MO} \rightarrow 0$ as $T_{ord} \rightarrow 0$ criterion [2]. This and alternative types of phase diagrams are schematically summarized in Fig. 1

The condition for $S_{MO} \rightarrow 0$ as $T_{ord} \rightarrow 0$ is fulfilled for systems represented by curve 'I'. The alternative types of magnetic phase diagrams can be recognized from the analysis of the specific heat jump at T_{ord} (ΔC_{MO}) [2]. Those ending at a finite temperature critical point because their ΔC_{MO} do not decrease sufficiently as T_{ord} decreases with a consequent bottleneck of S_{MO} at low temperature, label as 'II' in Fig. 1. Another group shows a transference of degrees of freedom from the MO phase to a non-magnetic component with their ΔC_m vanishing at finite temperature, represented by curve 'III' in Fig. 1.

Despite of the clear thermodynamic constraint (c.f. $S_{MO} \rightarrow 0$ when $T_{ord} \rightarrow 0$) for reaching a QCP the phase diagrams of real systems exhibit intrinsic differences with theoretical predictions. For example, their 2nd order magnetic phase boundaries at $T_{ord}(x)$ driven by a 'non-thermal' control parameter [3] like chemical potential, do not decrease according to the expected negative curvature down to $T \rightarrow 0$ because they deviate from that monotonous behavior at $T^{CR} \approx 2.5$ K [2]. This experimental observation

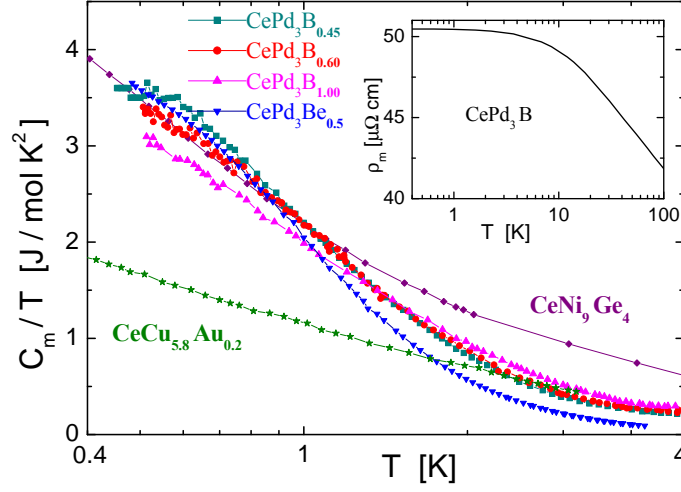


FIG. 2. (Color online) Low temperature specific heat of the heaviest fermions reported among Ce-lattice compounds in a log T representation. The well known heavy fermion (HF) $\text{CeCu}_{5.8}\text{Au}_{0.2}$ [9] is included for comparison. Inset: low temperature magnetic contribution to the electrical resistivity of CePd_3B after [7] in a logarithmic temperature scale.

invalidates misleading extrapolations of $T_{ord}(x) \rightarrow 0$ performed according to conventional criteria. A pre-critical region is identified below T^{CR} where the nature of the magnetic phase boundary undergoes significant modifications, like e.g. to become of 3rd order with a linear $T_{ord}(x)$ dependence. That 3rd order transition, identified from a jump in $\partial C_m / \partial T$ [4], is suppressed by moderate magnetic field without changing T_{ord} . Also an increasing remnant entropy (up to $0.4 R \ln 2$) is observed in this critical region as $T \rightarrow 0$ [5].

In this work we analyze a fourth group of Ce compounds (see arrow label as 'IV' in Fig. 1) which do not order magnetically because they behave as tuned at $T_{ord} = 0$. These systems collect the highest density of low energy excitations reflected in huge values of C_m/T at $T \rightarrow 0$. They represent an empirical upper limit for $\lim_{T \rightarrow 0} \partial S_m / \partial T = C_m/T$ because the systems showing magnetic order condense their magnetic degrees of freedom into the ordered phase between $T_{ord} > T > 0$.

II. EXPERIMENTAL RESULTS

A record high C_m/T value at low temperature was claimed for the compound CePd_3B more than two decades ago [6]. Despite of the lack of magnetic order down to $T = 0.5\text{K}$ there were, however, some indications for the onset of short range magnetic correlations from a broad maximum around $T = 0.7\text{K}$. A new series of samples were produced improving the annealing process which is reflected in a smoother $C_m(T)/T$ dependence around that temperature.

In Fig. 2 we present the experimental results on the new CePd_3B_x series with $x = 0.45, 0.60, 1$ and

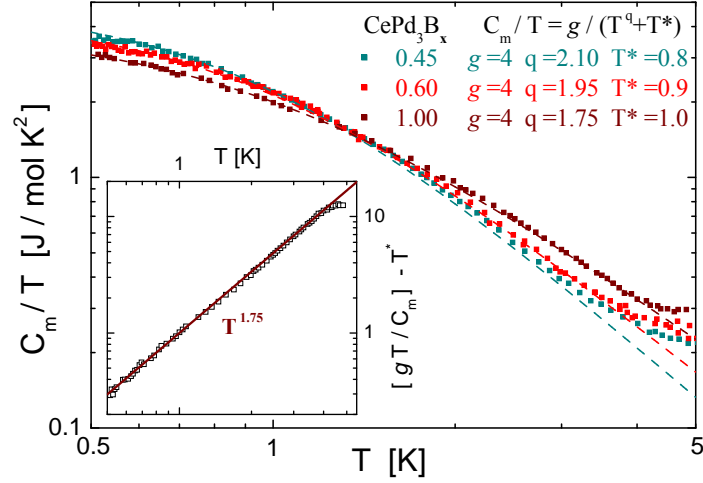


FIG. 3. (Color online) Fits for low temperature specific heat of CePd_3B_x compounds with a modified power law function. Inset: normalized inverse of C_m/T to extract the power law exponent for CePd_3B in a double logarithmic representation (see the text).

$\text{CePd}_3\text{Be}_{0.5}$ down to $T \approx 0.5\text{ K}$ in a logarithmic temperature scale. The magnetic contribution was obtained after subtracting the phonon component extracted from LaPd_3B [7]. For comparison, also CeNi_9Ge_4 [8] and $\text{CeCu}_{5.8}\text{Au}_{0.2}$ [9] compounds are included. The former shows the highest $C_m/T|_{\lim T \rightarrow 0}$ value reported at present among Ce-lattice intermetallics, whereas the later is the prototype of non-Fermi-liquids (NFL) accessing to QC regime with a logarithmic: $C_m/T \propto -\ln(T/T_0)$ dependence. CeNi_9Ge_4 was measured down to $\approx 50\text{ mK}$, where it reaches a record value of $C_m/T = 5.5\text{ J/molK}^2$. Such an extremely high value is associated to the contribution of the ground state and first excited crystal field (ECF) levels with a splitting comparable to the Kondo temperature $T_K = 10\text{ K}$ [8] which makes this system to be considered as an effective $N_{eff} > 2$ degenerated ground state (GS). Nevertheless, in Fig. 2 it can be seen that CePd_3B_x compounds reach similar C_m/T values for $0.5 \leq T \leq 1\text{ K}$ despite of their $N = 2$ GS character. Such a coincidence can be explained by the irrelevant Kondo effect in these cubic ternary compounds. From their magnetic properties one may distinguish between an effective $N_{eff} > 2$ GS and an actual $N = 4$. In the former two Kramer doublets with different giromagnetic factors (g_i) overlap in energy because the ECF splitting (Δ_I) compares with their level broadening (i.e. $\Delta_I/T_K \approx 1$). On the contrary, in the $N = 4$ case Kondo effect may be even absent like in BCC Ce-binary compounds [10].

Unlike $C_m/T \propto -\ln(T/T_0)$ dependence of $\text{CeCu}_{5.8}\text{Au}_{0.2}$, it is evident from Fig. 2 that CeNi_9Ge_4 and CePd_3B_x compounds obey a power law at low temperature. In order to check that behavior, we analyze in Fig. 3 such dependence for CePd_3B_x compounds using a modified power law $C_m/T = g/(T^q + T^*)$ [5], obtaining similar values of the exponent $q = 1.95 \pm 0.2$ and T^* values around 1 K for all of them.

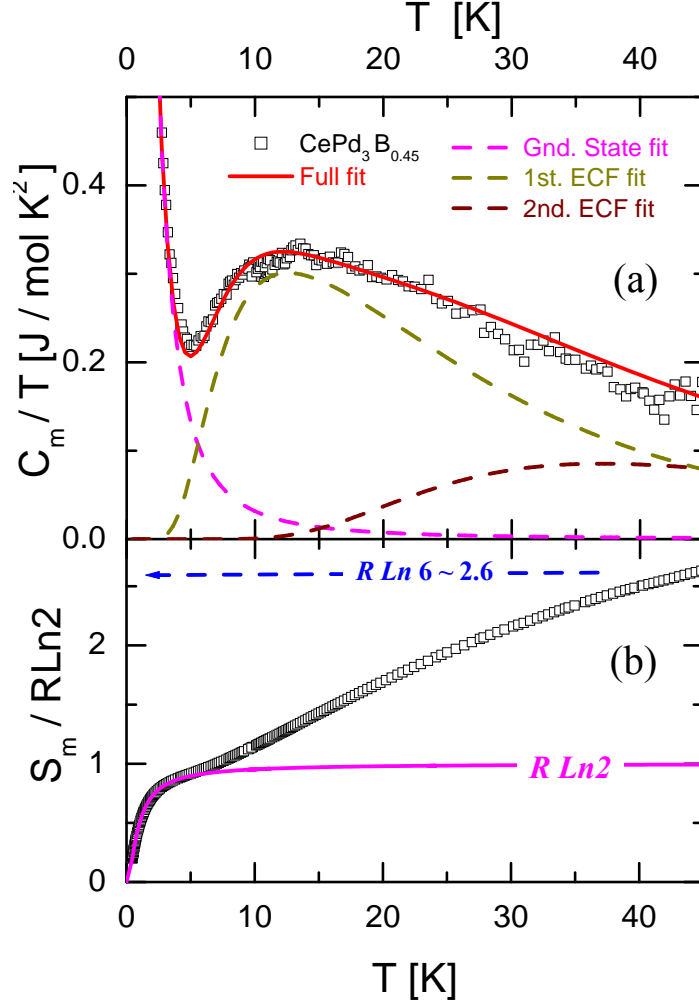


FIG. 4. (Color on line) (a) High temperature specific heat of $\text{CePd}_3\text{B}_{0.45}$ described accounting for the 1st. and 2nd. ECF doublets. (b) Thermal dependence of the magnetic entropy in $R \ln 2$ units.

The extrapolation down to $T = 0$ is given by $C_m/T|_{\lim T \rightarrow 0} = g/T^* = 4.5 \pm 0.5 \text{ J/molK}^2$. The accuracy of this description is evidenced in the inset of Fig. 3 where a double logarithmic representation allows to extract the exponent $q = 1.78$ for CePd_3B . Notice that the applied power law excludes spin or cluster glass behaviors because those cases follow a $C_m \propto 1/T^2$ dependence (i.e. $C_m/T \propto 1/T^3$) [11].

The $N = 2$ GS character of cubic CePd_3B_x compounds is confirmed by high temperature C_m measurements. Above $T \approx 5 \text{ K}$, the $C_m(T)/T$ results deviate from power law T dependencies because the first ECF level starts to contribute. In Fig. 4a we analyze that contribution on the high temperature $C_m(T)/T$ results from $\text{CePd}_3\text{B}_{0.45}$. A good overall fit is obtained accounting for the GS power law dependence and two Schottky anomalies originated in the contribution of two ECF doublets centered at $\Delta_I \approx 27 \text{ K}$ and $\Delta_{II} \approx 60 \text{ K}$ respectively. In this case, a level width corresponding to a T_K^I of $\approx 10 \text{ K}$ is required for

the first ECF doublet. Nevertheless, in comparison with CeNi_9Ge_4 , in this compound the $\Delta_I/T_K^I \approx 3$ ratio makes the GS to retain its actual $N = 2$ character. Such a difference is also reflected in the thermal dependence of the entropy (see Fig. 4b) which shows a tendency to a $S_m \rightarrow RLn2$ saturation around 7 K, prior the onset of the ECF levels contribution.

III. DISCUSSION

The basic thermodynamic implications of the $T \rightarrow 0$ physics regard the third law of thermodynamics which governs the $\lim_{T \rightarrow 0} \partial S / \partial T = C_m / T$. The conventional scenario for $\partial S / \partial T = 0$ corresponds to the case of a singlet, realized e.g. in a long-range-order GS associated to a positive curvature of $S_m(T)$ (i.e. $\partial^2 S_m / \partial T^2 > 0$) [12]. The case of $\partial S / \partial T \neq 0$ (with $\partial^2 S_m / \partial T^2 = 0$) has its simplest example in metallic systems whose conduction electrons are described as a Fermi gas. In this case, $\partial S / \partial T = \gamma$ defines the Sommerfeld coefficient typically in the range of a few [mJ/molK²]. Non magnetic Ce compounds in the intermediate valence regime, behave as Fermi liquids (FL) with $10 \leq \gamma \leq 100$ mJ/molK² values, whereas those with $100 \leq \gamma \leq 1$ J/molK² are recognized as HF. Within this group, one should distinguish between those showing FL and NFL character. The former keep their fermionic nature and form narrow bands of heavy quasi-particles of enhanced effective mass, typically exhibiting γ values up to 400 mJ/molK². The later present a dense spectrum of low energy excitations which not necessarily form a band. The $C_m(T)/T$ dependence typically obeys logarithmic or $a - b\sqrt{T}$ functions [15], in coincidence with an eventual Kondo-breakdown scenario [13]. Consequently, C_m/T cannot be identified with the canonical Sommerfeld γ coefficient because its temperature dependence differs from that of a FL since $C_m/T|_{\lim T \rightarrow 0} = \gamma * [1 - (T/T_K)^2]$ in the later [16].

A. $T \rightarrow 0$ divergencies

Even higher values are observed in a few very heavy fermion (VHF) ranging within $1 \leq C_m/T|_{\lim T \rightarrow 0} < 8$ J/molK², which do not show magnetic order down to the mK region of temperature. The analysis of their thermodynamic properties as $T \rightarrow 0$ is the main scope of this work. Typical thermal dependencies for these NFL systems are $-Ln(T/T_0)$ or T^{-q} [1, 17, 18], with the former not exceeding ≈ 3 J/molK² [9]. Both dependencies imply that $C_m/T|_{\lim T \rightarrow 0}$ keeps increasing because the density of low lying energy excitations grow continuously.

Since thermodynamic postulates and experimental evidences indicate that singularities at $T = 0$ and consequent negative $S_m(T \rightarrow 0)$ curvature ($\partial^2 S_m / \partial T^2 < 0$) are not physical, the question arises whether there is any upper limit for the $\partial S / \partial T = C_m / T$ slope. The experimental results collected in Fig. 2 for VHF Ce-lattice intermetallics suggest the existence of such upper limit, reached following the modified power law $C_m/T = g/(T^q + T^*)$.

Comparing characteristic energies of Ce compounds whose magnetically ordered phase fulfils the condition of $S_{MO} \rightarrow 0$ when $T_{ord} \rightarrow 0$ (described as case 'I' in ref. [2]), one finds that there is a significant coincidence in the scales of energy extracted from different types of analysis. In fact, the compounds fulfilling the mentioned condition show a clear change of regime at $T^{CR} = 2.5 \pm 0.3$ K [2], whereas in those systems which do not order magnetically the characteristic energy scale related to the deviation from a pure power law (i.e. the parameter T^* extracted from Fig. 3 fits) is around 1 K. If T^{CR} represents a threshold between thermal and quantum fluctuations dominated regions, the T^* value indicate that the deviation from the power law dependence may occur as an alternative to magnetic order.

The question arises concerning the reason why the Ce compounds collected in Fig. 3 do not order magnetically down to such a low temperature despite their robust Ce^{3+} moment. A common feature of these compounds is that the magnetic (Ce) lattice presents a subtle displacements from their strict periodic atomic positions frustrating the development of any long-range-order parameter. This possibility is supported by the large available volume of Ce atoms in CeNi_9Ge_4 and the effect of random distribution of B or Be interstitials in CePd_3X_x . Preliminary studies on a novel compound with the TiNiSi type structure showing very similar $C_m(T)$ dependence [20] confirms these observations.

As remarked in Ref.[13], in HF systems tuned by chemical pressure the coupling of electrons to lattice degrees of freedom can play a nontrivial role. In our case, due to the very low value of the characteristic energy scale, the local moments may form a spin-liquid state dominated by quantum fluctuations [13] without braking any symmetry (i.e. no phase transition is required). In fact, electrical resistivity of CePd_3B continuously increases at low temperature not showing any coherence effect [7], see the inset in Fig. 2. This scenario does not map a single impurity picture because, like La doped systems $(\text{Ce},\text{La})\text{Ni}_9\text{Ge}_4$ [8], $(\text{Ce}_{0.1}\text{La}_{0.9})\text{TiGe}$ [21] and $(\text{Ce}_{0.03}\text{La}_{0.97})\text{B}_6$ [22] which have quite high C_m/T values, they do not show evidences for $C_m/T \mid_{\lim T \rightarrow 0}$ saturation but a $-\ln(T/T_0)$ type dependence. Notably, the power law temperature dependence of Ce-lattice CeNi_9Ge_4 transforms into a $-\ln(T/T_0)$ one by La doping in $(\text{Ce},\text{La})\text{Ni}_9\text{Ge}_4$ [8]. The universality of a logarithmic type of temperature dependence is remarked by the existence of a single expression $C_m/t = -7.2\ln(t)$ [14] once the temperature is normalized by $t = T/T_0$, being T_0 a characteristic temperature of each system.

B. Entropy

In this subsection we will discuss the thermodynamic consequences of a $C_m/T = \partial S/\partial T$ upper limit on the thermal variation of the entropy at the $T \rightarrow 0$ limit. In Fig. 5a we have collected the low temperature $S_m(T)$ dependencies extracted from a number of $N = 2$ VHF Ce systems reaching $C_m/T \geq 3$ J/molK² values, independently of their temperature dependencies and whether they order or not. That figure includes $\text{CePd}_3\text{Be}_{0.5}$, two exemplary concentrations of the well known $\text{CeCu}_{6-x}\text{Au}_x$ [23] and $\text{CeCu}_{5.5}\text{Ag}_{0.5}$ [24]. Also diluted $(\text{Ce}_{0.1}\text{La}_{0.9})\text{TiGe}$ [21] is included to confirm that this limit is independent of lattice

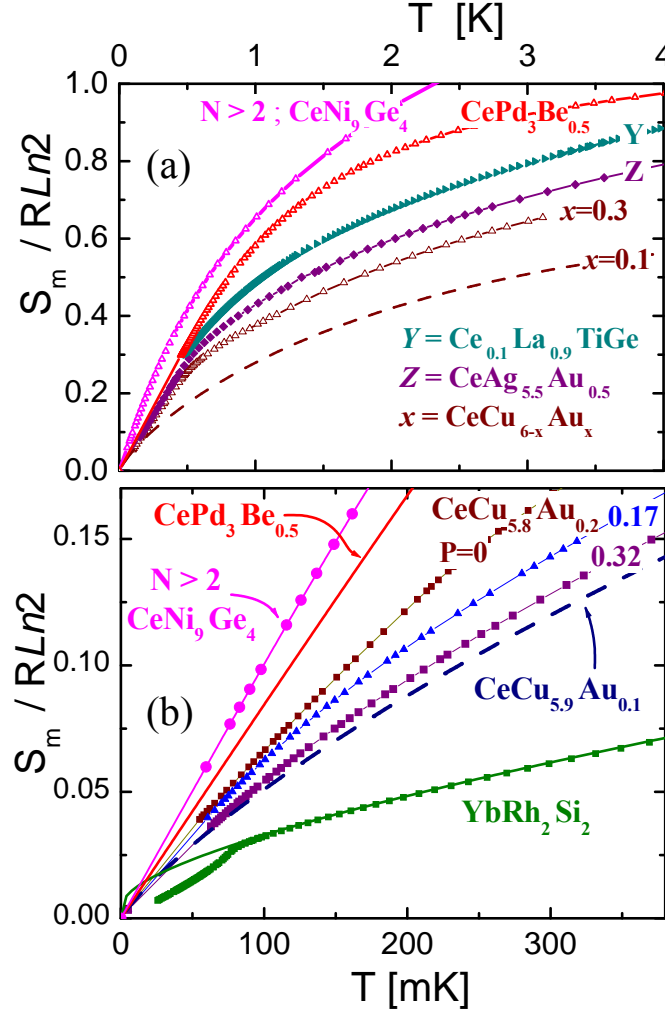


FIG. 5. (Color online) Examples of maximum slope of $S_m(T)$ for Cerium $N=2$ GS systems, (a) within the $0 < T < 4$ K range including $\text{CePd}_3\text{B}_{0.45}$, $\text{CeCu}_{6-x}\text{Au}_x$ [23], $(\text{Ce}_{0.1}\text{La}_{0.9})\text{TiGe}$ [21], $\text{CeAg}_{6-z}\text{Au}_z$ [24], and CeNi_9Ge_4 with $N_{eff} > 2$ [8]. (b) Detail of the $S_m(T)$ dependence within the mK range including: $\text{CeCu}_{5.9}\text{Au}_{0.1}$ (dashed line), $\text{CeCu}_{5.8}\text{Au}_{0.2}$ at different pressures (in GPa) and, for comparison, the power law fit of $\text{CePd}_3\text{Be}_{0.5}$ extrapolated from $T > 0.5$ K. The $N_{eff} > 2$ CeNi_9Ge_4 (after [8]) and the entropy evaluation from the well known YbRh_2Si_2 [26] are also included for a wider comparison.

configurations, and the entropy of CeNi_9Ge_4 [8] for comparison with a $N_{eff} > 2$ compound.

As it can be seen, experimental results suggest a sort of *entropy envelope curve* for $N = 2$ GS systems which is qualitatively represented by the $S_m(T)$ dependence of $\text{CePd}_3\text{Be}_{0.5}$. The highest $\partial S / \partial T$ is shown by the $N_{eff} > 2$ GS compound CeNi_9Ge_4 , which is overcome by Ce-diluted $(\text{Ce}_{0.03}\text{La}_{0.97})\text{B}_6$ [22] (not shown) which derives from the $N = 4$ GS compound CeB_6 with BCC structure. A closer analysis of the mK region is presented in Fig. 5b, with the *entropy envelope curve* of $\text{CePd}_3\text{B}_{0.45}$ extrapolated from the $T > 0.5$ K fit which shows a $C_m / T |_{\lim T \rightarrow 0} \approx 5 \text{ J/molK}^2$. Notably, Ce-lattice systems claimed

to diverge as $-Ln(T/T_0)$ fit into that limit. In fact, the highest HF system $CeCu_{6-x}Au_x$ shows a $C_m/T = -0.63Ln(T/5.27)$ dependence [23] which, at the extremely low temperature magnetic order of $T_N \approx 2$ mK [25], reaches the expected limit value of ≈ 5 J/molK².

For comparison with other Rare earth compounds, the thermal dependence of the entropy of $YbRh_2Si_2$ is included in Fig. 5b. Also in this case a first order transition occurs, at ≈ 80 mK [26], close to the *entropy envelope curve*, whereas the curve extrapolated from the paramagnetic state using the modified power law function would have exceed that value below about 40 mK. Similar situation occurs with the novel Yb compound $YbNi_4P_2$ [27] which also undergoes a first order transition at $T_C = 0.17$ K approaching the same *envelope curve*. In both cases $C_m/T |_{lim T \rightarrow 0} \approx 2$ J/molK², which is much smaller than 5 J/molK² reported for $YbCu_{5-x}Au_x$ [28] and the record one for $YbCo_2Zn_{20}$ of 7.8 J/molK² [29]. This compound does not order magnetically down to the mK range of temperature, but is considered to have a $N = 4$ GS [29].

IV. SUMMARY

Using selected specific heat results, we have analyzed the thermodynamic implications of the $T \rightarrow 0$ physics in VHF Ce compounds which do not order magnetically but exhibit extremely high $C_m/T |_{lim T \rightarrow 0}$ values. From these experimental information we have observed that: i) according to thermodynamic laws, $C_m(T)/T$ power law temperature dependencies tend to saturate, ii) the onset of that saturation occurs within the range of temperature dominated by quantum fluctuation (i.e. $T < 2$ K), which may lead to access to an exotic GS like e.g. a spin liquid one. iii) Alternatively to saturation the systems undergo magnetic transitions because of the entropy accumulation at very low temperature. iv) An empirical upper limit of $C_m/T |_{lim T \rightarrow 0} \approx 4.5$ J/molK² for Ce compounds with $N = 2$ GS is observed. Higher values are found in compounds with $N_{eff} > 2$ ground state. These experimental observations highlight the role of thermodynamic laws in the understanding of realistic $T \rightarrow 0$ physics, being specific heat and entropy the tools to distinguish between real and 'wished' candidates to quantum critical regime.

We conclude that thermodynamic laws and quantum critical mechanisms intervene simultaneously in the GS formation. Third law constraint on entropy accumulation at $T \rightarrow 0$ interdicts C_m/T singularities imposing an upper limit to the density of low lying quantum excitations. This constraint drives the systems to alternative GS through first order transitions (even at a few mK) or to the formation of exotic states. The access to a quantum critical point a $T \rightarrow 0$ seems to be limited by simple thermodynamic conditions before a technical limit of the cooling process occurs.

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